

FORM PTO-1390 (Modified) (REV 10-95)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER 200197US0XPCT
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 09/701376
INTERNATIONAL APPLICATION NO. PCT/JPO/001949	INTERNATIONAL FILING DATE 29 MARCH 2000	PRIORITY DATE CLAIMED 31 MARCH 1999		
TITLE OF INVENTION				
PROPYLENE-ETHYLENE BLOCK COPOLYMER COMPOSITION FOR AUTOMOBILE EXTERIOR PARTS				
APPLICANT(S) FOR DO/EO/US				
Yutaka KOBAYASHI, et al.				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
<ol style="list-style-type: none"> <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). <input type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). <input checked="" type="checkbox"/> has been transmitted by the International Bureau. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210). <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> have been transmitted by the International Bureau. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. <input checked="" type="checkbox"/> have not been made and will not be made. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). <input type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). 				
Items 13 to 18 below concern document(s) or information included:				
<ol style="list-style-type: none"> <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. <input type="checkbox"/> A substitute specification. <input type="checkbox"/> A change of power of attorney and/or address letter. <input type="checkbox"/> Certificate of Mailing by Express Mail <input checked="" type="checkbox"/> Other items: information: 				
Request for Consideration of Documents Cited in International Search Report Notice of Priority PCT/IB/304 PCT/IB/308				

U.S. APPLICATION NO. (IF KNOWN SEE 37 CFR 09/701376	INTERNATIONAL APPLICATION NO. PCT/JPO0/01949	ATTORNEY'S DOCKET NUMBER 200197USOXFCT		
20. The following fees are submitted. BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :		CALCULATIONS PTO USE ONLY		
<input checked="" type="checkbox"/> Search Report has been prepared by the EPO or JPO \$860.00 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) \$690.00 <input type="checkbox"/> No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) \$710.00 <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$1000.00 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$100.00				
ENTER APPROPRIATE BASIC FEE AMOUNT =		\$ 860.00		
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)).		□ 20 □ 30 0.00		
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	3 - 20 =	0	x \$18.00	0.00
Independent claims	1 - 3 =	0	x \$80.00	0.00
Multiple Dependent Claims (check if applicable).				□
TOTAL OF ABOVE CALCULATIONS =				\$ 860.00
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable).		□	0.00	
SUBTOTAL =				\$ 860.00
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)).		□ 20 □ 30 +	0.00	
TOTAL NATIONAL FEE =				\$ 860.00
Fee for recording the enclosed assignment (37 CFR 1.21(b)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).		□	0.00	
TOTAL FEES ENCLOSED =				\$ 860.00
		Amount to be: refunded	\$	
		charged	\$	
<input checked="" type="checkbox"/> A check in the amount of \$ 860.00 to cover the above fees is enclosed. <input type="checkbox"/> Please charge my Deposit Account No. in the amount of to cover the above fees. A duplicate copy of this sheet is enclosed. <input type="checkbox"/> The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. 15-0030 A duplicate copy of this sheet is enclosed.				
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.				
SEND ALL CORRESPONDENCE TO:				
 22850				
<i>Surinder Sachar</i> Surinder Sachar Registration No. 34,423				
SIGNATURE <hr/> <i>Surinder Sachar</i> NAME Norman F. Oblon <hr/> 24,618 REGISTRATION NUMBER <hr/> 11-30 00 DATE				

DESCRIPTION

PROPYLENE-ETHYLENE BLOCK COPOLYMER COMPOSITION FOR
AUTOMOBILE EXTERIOR PARTS

5

TECHNICAL FIELD

The present invention relates to a propylene-ethylene block copolymer composition for use in automobile exterior parts such as bumper fascias, splash shields and side moldings, and more particularly to a propylene-ethylene block copolymer composition having a sufficient mechanical strength required for automobile exterior parts, which is produced at a low cost without additional steps for blending other rubber components or the like.

BACKGROUND ARTS

15 Hitherto, automobile exterior parts such as bumper fascias, splash shields and side moldings have been produced from polypropylene-based composite materials prepared by blending polypropylene with other components such as rubbers and talc. However, the production of these composite materials requires additional steps for blending the other
20 components such as rubbers and talc with the polypropylene. For this reason, there has been increasing demand for developing a composition capable of being molded into automobile exterior parts by itself without blending other components, and many attempts have been made.

For example, Chikashi Okayama, "Chemical Economy", pp. 8-15
25 (October, 1996) discloses a gas-phase polymerization process for producing a polypropylene block copolymer in which the solid structure of the product is micro-controlled by carrying out the polymerization in the presence of a specific catalyst. However, the resultant composition, which contains no inorganic materials and has a specific gravity of 0.89 or less, is unsatisfactory in stiffness

and mechanical strength required for automobile exterior parts since the flexural modulus is as low as 700 MPa and the -30°C Izod impact strength is as low as 120 kJ/m² at most.

5 **DISCLOSURE OF THE INVENTION**

The present invention has been made to solve the above problems. An object of the present invention is to provide a propylene-ethylene block copolymer composition for automobile exterior parts, which has sufficient stiffness and mechanical strength required for the automobile exterior parts
10 and is produced at low cost without additional steps for blending other components such as rubbers.

As a result of extensive researches in view of the above object, the inventors have found that the object is accomplished by blending a limited amount of a nucleating agent comprising methylenebis(2,4-di-t-butylphenol)
15 acid sodium phosphate with a propylene-ethylene block copolymer upon granulation thereof to produce a composition having specific structure and properties. The present invention has been completed based on this finding.

Namely, the propylene-ethylene block copolymer composition of the present invention is a composition for use in producing automobile exterior parts, comprising a propylene-ethylene block copolymer and a nucleating agent comprising methylenebis(2,4-di-t-butylphenol) acid sodium phosphate which is blended with the propylene-ethylene block copolymer in an amount of 300 to 2,000 ppm upon granulation thereof, and having (a) a melt flow rate (MFR) of 10 to 18 g/10 min when measured at 230°C under a load of 2.16 kg (21.2N); (b)
25 an ordinary temperature xylene insoluble having a stereoregularity index [mmmm] fraction of 98.9% or higher when measured by ¹³C-NMR; and (c) an ordinary temperature xylene soluble characterized by: (c-1) having a content of 22 to 28% by weight; (c-2) comprising only a single component with respect to a relaxation time T1 measured by pulse NMR; and (c-3) satisfying the following

formula (I):

$$y \leq 0.0014x^3 - 0.0897x^2 - 1.0593x + 231.6 \quad (I)$$

wherein x is an ethylene content (% by weight) measured by ^{13}C -NMR and y is the relaxation time T1 (msec) measured by pulse NMR.

5 The propylene-ethylene block copolymer composition of the present invention has (d) a flexural modulus of 1,000 to 1,500 MPa; (e) a -30°C Izod impact strength of 6 to 8 kJ/m²; and (f) a tensile elongation of 200% or higher.

The propylene-ethylene block copolymer composition of the present invention is suitably used for the production of automobile exterior parts such 10 as bumper fascias, splash shields and side moldings.

PREFERRED EMBODIMENT TO CARRY OUT THE INVENTION

The propylene-ethylene block copolymer composition of the present invention comprises a polypropylene part, a propylene-ethylene copolymer part 15 and a polyethylene part, and (a) contains 300 to 2000 ppm of methylenebis(2,4-di-t-butylphenol) acid sodium phosphate (NA11) as a nucleating agent which is added when the propylene-ethylene block copolymer produced in the form of powder is granulated. When the addition amount is less than 300 ppm, automobile exterior parts molded from the composition are insufficient in 20 stiffness such as flexural modulus. The addition of the nucleating agent more than 2,000 ppm produces no improvement in stiffness.

The propylene-ethylene block copolymer composition of the present invention has a melt flow rate (MFR) of 10 to 18 g/10 min, preferably 12 to 16 g/10 min when measured at 230°C under a load of 2.16 kg (21.2N). When the 25 melt flow rate of the composition is less than 10 g/10 min, the composition is difficult to be molded due to its insufficient fluidity. When the melt flow rate is more than 18 g/10 min, the resultant molded products are insufficient in mechanical strength such as tensile elongation.

The propylene-ethylene block copolymer composition of the present

invention contains (b) an ordinary temperature xylene insoluble having a stereoregularity index [mmmm] fraction of 98.9% or higher when measured by ^{13}C -NMR.

The ordinary temperature xylene insoluble substantially comprises the propylene homopolymer part. The stereoregularity index [mmmm] fraction, referred to also as an isotactic pentad fraction, means a percentage of the number of meso-bonded pentad sequences of propylene monomer units to the total number of propylene monomer units present in polypropylene molecular chain. Therefore, a polypropylene becomes highly isotactic with increasing isotactic pentad fraction. When the stereoregularity index [mmmm] fraction is less than 98.9%, the composition is insufficient in stiffness such as flexural modulus.

The propylene-ethylene block copolymer composition of the present invention contains (c1) an ordinary temperature xylene soluble in an amount of 22 to 28% by weight.

The ordinary temperature xylene soluble substantially comprises the ethylene-propylene copolymer part, and contains no crystalline polyethylene. When the content of the xylene soluble is less than 22% by weight, the composition is unsatisfactory in low-temperature impact resistance. When the content of the xylene soluble is more than 28% by weight, the composition is unsatisfactory in stiffness such as flexural modulus and deteriorated in surface hardness and heat resistance.

The ordinary temperature xylene soluble (c2) comprises only a single component with respect to the relaxation time T1 measured by pulse NMR, and (c3) satisfying the following formula (I):

$$y \leq 0.0014x^3 - 0.0897x^2 - 1.0593x + 231.6 \quad (\text{I})$$

wherein x is an ethylene content (% by weight) measured by ^{13}C -NMR and y is the relaxation time T1 (msec) measured by pulse NMR.

When an electromagnetic radiation pulse of a specific frequency is

applied to the ordinary temperature xylene soluble, the nuclear magnetic moment of the specimen is excited and then returns to the initial state. The time taken to return to the initial state is called as the relaxation time T1. The relaxation time T1 measured by pulse NMR indicates a magnitude of molecular motion of the specimen. A short relaxation time T1 of the specimen corresponds to a low frequency range of the molecular motion. The fact that a specimen having a short relaxation time T1 exhibits a high impact resistance suggests that the time scale for the impact test is close to the above frequency range.

- 5 10 Also, the fact that the xylene soluble contains only a single component with respect to the relaxation time T1 means that the xylene soluble shows properties close to those of a uniform component. That is, the satisfaction of the formula (I) shows that the ethylene-propylene copolymer part, irrespective of its inclusion of ethylene, is low in frequency range of molecular motion and
- 15 constituted by a rubber-like elastomer which substantially comprises a single component.

The relaxation time T1 of the ordinary temperature xylene soluble is determined by pulse NMR as described below.

- Based on the results of pulse NMR measurement, the values of $\ln\{M(\infty) - M(0)\}$ are plotted as a function of the variable time τ (irradiation interval between 180° pulse and 90° pulse) taking the intensity of signals after applying 90° pulse as $M(\tau)$. The plots produce a line which slopes down toward the right, and the relaxation time T1 is given by the absolute value of the reciprocal of the slope. A steeper slope of the line means a smaller relaxation time T1 and
- 20 25 immediate decay in shorter period of time. If plots produce various lines with different slopes, it means that the xylene soluble comprises a plurality of components with respect to the relaxation time T1. Therefore, in order that the xylene soluble comprises only a single component with respect to the relaxation time T1, the plots are required to form a single line.

The propylene-ethylene block copolymer composition of the present invention is deteriorated in impact strength if the ordinary temperature xylene soluble fails to comprise only a single component with respect to the relaxation time T1 measured by pulse NMR. Further, if the formula (I) is not satisfied, 5 the composition is also deteriorated in impact strength.

The propylene-ethylene block copolymer composition of the present invention is required to have (d) a flexural modulus of 1,000 to 1,500 MPa, (e) a -30°C Izod impact strength of 6 to 8 kJ/m², and (f) a tensile elongation of 200% or higher.

- 10 When the flexural modulus is less than 1,000 MPa, the composition is too soft to retain the shapes of the automobile exterior parts. When the flexural modulus is more than 1,500 MPa, the composition is deteriorated in low-temperature impact resistance. When the -30°C Izod impact strength is less than 6 kJ/m², the composition is poor in impact resistance, so that the resultant 15 automobile exterior parts tend to be broken upon collision. When the -30°C Izod impact strength is more than 8 kJ/m², the flexural modulus of the composition is reduced. When the tensile elongation of the composition is less than 200%, the resultant automobile exterior parts tend to be broken upon collision.
- 20 As described above, since a nucleating agent comprising methylenebis(2,4-di-t-butylphenol) acid sodium phosphate is blended in a limited amount, the propylene-ethylene block copolymer composition of the present invention satisfies the above requirements (a) to (f), i.e., the composition is highly balanced between stiffness such as flexural modulus and 25 mechanical strength such as impact strength. Therefore, the composition of the present invention is suitably applied to the automobile exterior parts without adding other rubber components or the like.

[Production of propylene-ethylene block copolymer composition]

The propylene-ethylene block copolymer composition of the present

invention may be produced by any suitable methods without particular limitations as long as the obtained composition satisfies the above requirements. In order to produce a polymer containing a high-stereoregular propylene homopolymer and having an excellent copolymerizability with

5 ethylene especially in a high molecular weight corresponding to MFR of about 12 to 16 g/10 min, the use of Ziegler catalysts rather than metallocene catalysts is preferred. More specifically, the polymerization may be carried out in the presence of, for example, a catalyst comprising (A) a solid catalyst component composed of (a) a titanium compound, (b) a magnesium compound, (c) an

10 electron donor and, if required, (d) a silicon compound; (B) an organoaluminum compound; and (C) an electron donating compound as a third component. In particular, the solid catalyst component usable in the present invention may be produced by contacting the magnesium compound and the titanium compound at 120 to 150°C in the presence of the electron donating compound and, if

15 required, the silicon compound, and then rinsing the contacted product with an inert solvent at 100 to 150°C.

In the present invention, the obtained polymer may be subjected to post-treatments by ordinary methods. Specifically, in the gas-phase polymerization, a nitrogen stream may be passed through a powdery polymer taken out of a polymerization vessel to remove propylene, ethylene, etc. in the powdery polymer. Alternatively, the obtained polymer may be pelletized using an extruder, if desired. Upon the extrusion, a slight amount of water, alcohol or the like may be added to the polymer to completely deactivate the remaining catalyst. In the bulk polymerization, the polymer taken out of a

20 polymerization vessel may be pelletized after completely removing remaining monomers.

Upon the pelletization, methylenebis(2,4-di-t-butylphenol) acid sodium phosphate as the nucleating agent is added to the obtained polymer in an amount of 300 to 2,000 ppm.

The present invention will be described in more detail with reference to the following examples, but these examples are not intended to limit the present invention thereto.

The analysis of structure and the measurement of properties of the 5 propylene-ethylene block copolymer composition were conducted by the following methods.

(1) Melt flow rate (MFR)

Measured at 230°C under a load of 2.16 kg (21.2N) according to JIS K7210.

10 (2) Ordinary temperature xylene soluble and xylene insoluble

The ordinary temperature (25°C) xylene soluble and xylene insoluble were obtained as follows.

- (a) Into a 1,000-ml round bottom flask, an accurately weighed sample (5 ± 0.05 g) was charged. After charging 1 ± 0.05 g of BHT (antioxidant), 700 ± 10 15 ml of p-xylene and a rotor were placed in the flask.
- (b) After fitting a condenser to the round bottom flask, the flask was heated for 120 ± 30 minutes in an oil bath maintained at $140 \pm 5^\circ\text{C}$ under stirring, thereby dissolving the sample into p-xylene.
- (c) The contents of the flask were poured into a 1,000-ml beaker. The 20 solution in the beaker was allowed to stand (for 8 hours or longer) under agitating by a stirrer until the solution was cooled to room temperature (25°C). Thereafter, the solution was filtered through a wire mesh to remove the precipitates.
- (d) The filtrate was filtered through a filter paper and then poured into 25 2,000 \pm 100 ml of methanol in a 3,000-ml beaker. The resultant mixture was allowed to stand at room temperature (25°C) for 2 hours or more while stirring with a stirrer.
- (e) Thereafter, the precipitates were separated by filtration through a wire mesh, air-dried for 5 hours or more and then vacuum-dried at $100 \pm 5^\circ\text{C}$ for 240

to 270 minutes in a vacuum dryer to recover a 25°C xylene soluble.

(f) The precipitates separated by filtration through the wire mesh in the step (c) were re-dissolved in p-xylene in the same manner as in the steps (a) and (b). The still hot solution was rapidly poured into 2,000 ± 100 ml of methanol

5 in a 3,000-ml beaker, agitated by a stirrer for 2 hours or more, and then allowed to stand at room temperature (25°C) over night.

(g) Thereafter, the precipitates were separated by filtration though a wire mesh, air-dried for 5 hours or more and then vacuum-dried at 100 ± 5°C for 240 to 270 minutes in a vacuum dryer to recover a 25°C xylene insoluble.

10 The content (w) of the 25°C xylene soluble is represented by the formula:

$$w \text{ (% by weight)} = 100 \times C/A$$

wherein A is a weight (g) of the sample and C is a weight (g) of the xylene soluble recovered in the step (e). The content of the xylene insoluble is represented by (100 - w) % by weight.

15 (3) Ethylene content of ordinary temperature xylene soluble measured by ^{13}C -NMR

In an NMR sample tube, 220 mg of a sample were placed and then 3 ml of a mixed solvent of 1,2,4-trichlorobenzene/heavy benzene (90/10 by volume) were added. After sealing the tube with a cap, the sample was dissolved in the 20 solvent at 130°C, and the resultant uniform solution was subjected to ^{13}C -NMR under the following conditions.

Apparatus used: JNM-EX400 manufactured by Nihon Denshi Co., Ltd.

Pulse width: 9 μs (45°)

Pulse interval: 4 seconds

25 Spectrum width: 20,000 Hz

Measuring temperature: 130°C

Number of integration: 1,000 to 10,000

The ethylene unit content (z) of the 25°C xylene soluble and the ethylene unit content (y) of the 25°C xylene insoluble were determined by the following

methods.

The triad sequence fractions (mol%) of ethylene (E) and propylene (P) were calculated from the following formulae, using the intensities of seven peaks present in a region of 35 to 21 ppm (chemical shift from

- 5 tetramethylsilane (TMS) standard) of a ^{13}C -NMR spectrum of the sample.

$$f_{EPE} = [K(T\delta\delta)/T] \times 100$$

$$f_{FPE} = [K(T\beta\delta)/T] \times 100$$

$$f_{EEE} = [K(S\gamma\delta)/4T + K(S\delta\delta)/2T] \times 100$$

$$f_{PPP} = [K(T\beta\beta)/T] \times 100$$

10 $f_{PEE} = [K(S\beta\delta)/T] \times 100$

$$f_{PEP} = [K(S\beta\beta)/T] \times 100$$

wherein $T = K(T\delta\delta) + K(T\beta\delta) + K(S\gamma\delta)/4 + K(S\delta\delta)/2 + K(T\beta\beta) + K(S\beta\delta) + K(S\beta\beta)$.

In the above formulae, for example, f_{EPE} is EPE triad sequence fraction (mol%), and $K(T\delta\delta)$ is an integrated intensity of the peaks assigned to $T\delta\delta$ carbon.

The ethylene unit content (%) by weight) is calculated from the following formula using the above triad sequence fractions.

$$\begin{aligned} \text{Ethylene unit content (\% by weight)} &= 28\{3f_{EEE} + 2(f_{PEE} + f_{EPE}) + f_{FPE} + f_{PEP}\} \\ &\times 100 / [28\{3f_{EEE} + 2(f_{PEE} + f_{EPE}) + f_{FPE} + f_{PEP}\} + 42\{3f_{PPP} + 2(f_{FPE} + f_{PEP}) + f_{EPE} + \\ &20 f_{PEE}\}] \end{aligned}$$

(4) Stereoregularity index of ordinary temperature xylene insoluble measured by ^{13}C -NMR

In a ^{13}C -NMR spectrum of the 25°C xylene insoluble, the signal of methyl carbon is observed as nine split peaks of mmmm, mmmr, rmmr, mmrr, mmrm + rrmr, rrrm, rrrr, mrrr and mrrm over a wide range from low magnetic field to high magnetic field due to the influence of stereoregularity. Of these nine peaks, the six peaks with higher intensities, i.e., mmmm, mmmr, mmrr, mmrm + rrmr, rrrr and mrrm are used to calculate the stereoregularity index of the xylene insoluble according to the following formula:

$$\text{Stereoregularity index (\%)} = \frac{L_{\text{mmmm}}}{L_{\text{mmmm}} + L_{\text{mmmr}} + L_{\text{mmrr}} + L_{\text{mmrm}} + L_{\text{rrrr}} + L_{\text{rrrm}}} \times 100$$

wherein L_{mmmm} , L_{mmmr} , L_{mmrr} , $L_{\text{mmrm}} + L_{\text{rrrr}}$, L_{rrrr} and L_{rrrm} represent peak heights from the base line of mmmm, mmmr, mmrr, mmrm + rrrr, rrrr and mrrm

- 5 peaks of a ^{13}C -NMR spectrum.

Since, the peak of mmmm is constituted by a plurality of discrete points having different chemical shifts and different peak heights, and the peak of mmmr appears on the tailing of the main peak of mmmm, the heights of these peaks from the base line are corrected by ordinary methods.

- 10 10 (5) Relaxation time T1 of ordinary temperature xylene soluble measured by pulse NMR

The relaxation time T1 is a time constant of a longitudinal magnetization recovery, and is determined most generally by an inversion recovery method ($180^\circ - \tau - 90^\circ$ pulse method). When $\theta = 180^\circ$ pulse is applied to a sample at the 15 time $t = 0$, the magnetization in the $-z'$ -axis direction is generated, and then the magnetization begins to return to thermal equilibrium value M_0 . After the elapse of time τ , when 90° pulse is applied, the magnetization is rotated to the y' -axis direction to create FID signals in proportion to the magnitude of magnetization. By continuously changing τ , a recovery curve of signal 20 intensity $M(\tau)$ is obtained. According to Bloch equation, the longitudinal magnetization recovery under the initial condition of $M(0) = -M_0$ is expressed by the following formula:

$$M = M_0 \{1 - 2\exp(-\tau/T_1)\}$$

- Further, in practice, using the relationship of $\ln\{M(\infty) - M(\tau)\} = 25 \ln\{2M(\infty)\} - \tau/T_1$, the relaxation time T1 is determined from the slope of line obtained by plotting $\ln\{M(\infty) - M(\tau)\}$ vs. τ .

Meanwhile, upon conducting a series of measurements including repeated tests for integration, the subsequent series of application of pulse should be delayed until the magnetization fully returns to thermal equilibrium

state. Since the delay time was at least 5T1 (99.3% return for the period of 5T1), the value M(τ) satisfying $\tau > 5T1$ was adopted as M(∞).

The relaxation time T1 was measured using a pulse NMR apparatus CXP-90 manufactured by Bruker Co., Ltd. under the following conditions:

5 Measuring nucleus: hydrogen nucleus

Measuring frequency: 90 MHz

Measuring temperature: 30°C

Measuring method: Inversion recovery method (180°- τ -90° pulse method)

10 180°: 180° pulse

90°: 90° pulse

τ : variable time

90° pulse width: 2.3 to 2.4 μ sec

(6) Flexural modulus

15 Measured according to ASTM D790.

(7) Izod impact strength

Measured according to ASTM D256 at -30°C using a notched injection-molded piece.

(8) Tensile elongation

20 Measured according to ASTM D638.

[Preparation of polymerization catalyst]

(1) Preparation of catalyst A

(Preparation of solid catalyst component)

A five-liter three-necked flask equipped with a stirrer which had been 25 purged with nitrogen, was charged with 160 g of diethoxy magnesium and further with 600 ml of dehydrated octane. The contents of the flask were heated to 40°C and then mixed with 24 ml of silicon tetrachloride. The resultant mixture was stirred for 20 minutes, and mixed with 16 ml of dibutyl phthalate. The obtained solution was heated to 80°C, followed by the dropwise

addition of 770 ml of titanium tetrachloride through a funnel. The contact of the reactants was carried out at an internal temperature of 125°C for 2 hours. Then, the stirring was stopped to allow the solid product to sediment, and a supernatant was discarded. After mixed with 100 ml of dehydrated octane,

5 the solid product was heated to 125°C under stirring and maintained at that temperature for one minute. Then the stirring was stopped to allow the solid product to sediment, and a supernatant was discarded. The above washing procedure was repeated seven times. The solid product was further contacted with 1,220 ml of titanium tetrachloride at an inner temperature of 125°C for 2

10 hours. Then, the solid product were further washed with dehydrated octane of 125°C six times to obtain a solid catalyst component.

(Pre-polymerization)

An one-liter three-necked flask equipped with a stirrer which had been purged with nitrogen, was charged with 48 g of the solid catalyst component

15 and then with 400 ml of dehydrated heptane. The contents of the flask were heated to 40°C and then added with 2.0 ml of triethylaluminum and 6.3 ml of dicyclopentyldimethoxysilane. The reaction was conducted by passing propylene gas through the resultant mixture under ordinary pressure for 2 hours. Thereafter, the solid component was thoroughly washed with

20 dehydrated heptane to obtain a catalyst A.

(2) Preparation of catalyst B

(Preparation of solid catalyst component)

A five-liter three-necked flask equipped with a stirrer which had been purged with nitrogen, was charged with 160 g of diethoxy magnesium and

25 further with 600 ml of dehydrated heptane. The contents of the flask were heated to 40°C and then mixed with 24 ml of silicon tetrachloride. The resultant mixture was stirred for 20 minutes, and added with 25 ml of dibutyl phthalate. The obtained solution was heated to 80°C, followed by the dropwise addition of 770 ml of titanium tetrachloride through a funnel. The contact of

the reactants was carried out at an internal temperature of 110°C for 2 hours. Then, the solid product was washed with dehydrated heptane of 90°C seven times, and then contacted with 1,220 ml of titanium tetrachloride at an inner temperature of 110°C for 2 hours. Then, the solid product was further washed 5 with dehydrated heptane of 60°C six times to obtain a solid catalyst component. (Pre-polymerization)

An one-liter three-necked flask equipped with a stirrer which had been purged with nitrogen, was charged with 48 g of the solid catalyst component and then with 400 ml of dehydrated heptane. The contents of the flask were 10 heated to 40°C and then added with 2.0 ml of triethylaluminum and 6.3 ml of dicyclopentyldimethoxysilane. The reaction was conducted by passing propylene gas through the resultant mixture under ordinary pressure for 2 hours. Thereafter, the solid component was thoroughly washed with dehydrated heptane to obtain a catalyst B.

15 (3) Preparation of catalyst C
(Preparation of solid catalyst component)

A five-liter three-necked flask equipped with a stirrer which had been purged with nitrogen, was charged with 160 g of diethoxy magnesium and further with 600 ml of dehydrated heptane. The contents of the flask were 20 heated to 40°C and then mixed with 24 ml of silicon tetrachloride. The resultant mixture was stirred for 20 minutes, and added with 23 ml of diethyl phthalate. The obtained solution was heated to 80°C, followed by the dropwise addition of 770 ml of titanium tetrachloride through a funnel. The contact of the reactants was carried out at an internal temperature of 110°C for 2 hours. 25 Then, the solid product was washed with dehydrated heptane of 90°C seven times, and then contacted with 1,220 ml of titanium tetrachloride at an inner temperature of 110°C for 2 hours. Then, the solid product was further washed with dehydrated heptane of 90°C six times to obtain a solid catalyst component. (Pre-polymerization)

- An one-liter three-necked flask equipped with a stirrer which had been purged with nitrogen, was charged with 48 g of the solid catalyst component and then with 400 ml of dehydrated heptane. The contents of the flask were added with 2.7 ml of triethylaluminum and 2 ml of
- 5 cyclohexylmethyldimethoxysilane, while kept at 10°C. The reaction was conducted by passing propylene gas through the resultant mixture under ordinary pressure for 2 hours. Thereafter, the solid component was thoroughly washed with dehydrated heptane to obtain a catalyst C.
- EXAMPLE 1
- 10 A five-liter stainless-steel autoclave equipped with a stirrer was sufficiently dried with nitrogen and then purged with propylene gas. While keeping the autoclave at 70°C, the inner pressure of the autoclave was increased to 0.05 MPa by introducing propylene gas. Then, hydrogen gas was introduced into the autoclave until the inner pressure of the autoclave reached
- 15 0.7 MPa, and then the inner pressure was gradually increased to 2.8 MPa by introducing propylene gas. Then, 20 ml of heptane, 4 mmol of triethyl aluminum, 1 mmol of dicyclopentyldimethoxysilane and 0.02 mmol of the catalyst A were placed in a 60-ml catalyst feed pipe which had been purged with a nitrogen gas and the mixture was added into the autoclave where the
- 20 polymerization was conducted for 60 minutes to produce a propylene homopolymer. Then, a mixed gas of ethylene/propylene in a molar ratio of 3.5 : 6.5 was introduced into the autoclave until the inner pressure reached 0.12 MPa. While keeping at 70°C under 1 MPa, propylene-ethylene copolymerization was conducted for 45 minutes. Thereafter, the autoclave was
- 25 evacuated to atmospheric pressure and the inner temperature of the autoclave was decreased to ordinary temperature. Then, the autoclave was opened to recover a resultant polymer powder.

The obtained polymer powder was intimately mixed with 1,000 ppm of calcium stearate as a neutralizing agent, 500 ppm of DHT-4A (magnesium-

- aluminum-hydroxide-carbonate-hydrate produced by Kyowa Kagaku Kogyo Co., Ltd.), 750 ppm of P-EPQ as an antioxidant (tetrakis(2,4-di-tert-butylphenyl-4,4-bisphenylenediphosphonite) produced by Asahi Denka Kogyo Co., Ltd.), 1,500 ppm of INGANOX1010 (phenol-based antioxidant produced by Ciba
- 5 Speciality Chemicals Co., Ltd.) and 1,200 ppm of NA11 as a nucleating agent (methylenebis(2,4-di-t-butylphenol) acid sodium phosphate produced by Asahi Denka Kogyo Co., Ltd.). The resultant mixture was melt-kneaded and granulated using a 20mmφ single screw extruder, thereby obtaining pellets of a propylene-ethylene block copolymer composition. A part of the pellets was
10 subjected to the structural analysis, and the remainder of the pellets was injection-molded to prepare various test pieces which were then subjected to measurement of the physical properties. The results are shown in Table 1.

EXAMPLE 2

Pellets of propylene-ethylene block copolymer composition were produced
15 by the same method as in Example 1 except that the propylene-ethylene copolymerization was conducted for 60 minutes and the amount of the nucleating agent NA11 was changed to 2,000 ppm. The structural analysis and measurements of physical properties were carried out by the same method as in Example 1. The results are shown in Table 1.

EXAMPLE 3

Pellets of propylene-ethylene block copolymer composition were produced by the same method as in Example 1 except that the molar ratio of ethylene /propylene was changed to 4.5 : 5.5. The structural analysis and measurements of physical properties were carried out by the same method as in
25 Example 1. The results are shown in Table 1.

EXAMPLE 4

Pellets of propylene-ethylene block copolymer composition were produced by the same method as in Example 1 except that the molar ratio of ethylene/propylene was changed to 5.5 : 4.5, and the amount of the nucleating

agent NA11 was changed to 800 ppm. The structural analysis and measurements of physical properties were carried out by the same method as in Example 1. The results are shown in Table 1.

EXAMPLE 5

5 Pellets of propylene-ethylene block copolymer composition were produced by the same method as in Example 1 except that the ethylene/propylene mixed gas was introduced until the inner pressure of the autoclave reached 0.9 MPa. The structural analysis and measurements of physical properties were carried out by the same method as in Example 1. The results are shown in Table 1.

10 COMPARATIVE EXAMPLE 1

Pellets of propylene-ethylene block copolymer composition were produced by the same method as in Example 2 except that cyclohexylmethyldimethoxysilane was used instead of dicyclopentyldimethoxysilane, the catalyst C was used instead of the catalyst A, 15 and the use of the nucleating agent NA11 was omitted. The structural analysis and measurements of physical properties were carried out by the same method as in Example 1. The results are shown in Table 2.

COMPARATIVE EXAMPLE 2

Pellets of propylene-ethylene block copolymer composition were produced 20 by the same method as in Example 2 except that cyclohexylmethyldimethoxysilane was used instead of dicyclopentyldimethoxysilane, and the catalyst C was used instead of the catalyst A. The structural analysis and measurements of physical properties were carried out by the same method as in Example 1. The results are shown 25 in Table 2.

COMPARATIVE EXAMPLE 3

Pellets of propylene-ethylene block copolymer composition were prepared by the same method as in Example 2 except that hydrogen gas was introduced into the autoclave until the inner pressure reached 0.55 MPa, the catalyst B

- was used instead of the catalyst A, a mixed gas of ethylene/propylene (molar ratio: 1 : 2) was introduced into the autoclave until the inner pressure reached 0.1 MPa, and the use of the nucleating agent NA11 was omitted. The structural analysis and measurements of physical properties were carried out 5 by the same method as in Example 1. The results are shown in Table 2.

COMPARATIVE EXAMPLE 4

Pellets of propylene-ethylene block copolymer composition were prepared by the same method as in Example 2 except that the mixed gas of ethylene/propylene was introduced into the autoclave until the inner pressure 10 reached 0.15 MPa, the propylene-ethylene copolymerization was conducted for 80 minutes, and the use of the nucleating agent NA11 was omitted. The structural analysis and measurements of physical properties were carried out

by the same method as in Example 1. The results are shown in Table 2.

Table 1

	Examples				
	1	2	3	4	5
Amount of nucleating agent (ppm)	1,200	2,000	1,200	800	1,200
MFR (g/10 min)	12.7	12.0	12.8	11.4	10.5
Stereoregularity index of ordinary temperature xylene insoluble (%)	99.1	99.1	99.1	99.0	99.1
Content of ordinary temperature xylene soluble (wt. %)	22.0	25.1	22.1	25.7	22.0
Relaxation time (T1) γ (msec)	146	147	140	134	146
Value of right side of formula (1)	149.2	149.5	140.2	136.2	149.2
Flexural modulus (MPa)	1,150	1,150	1,180	1,050	1,180
-30°C Izod impact strength (kJ/m ²)	6.5	7.2	6.6	8.0	7.1
Tensile elongation (%)	≥500	250	200	300	≥500

Table 2

	<u>Comparative Examples</u>			
	1	2	3	4
Amount of nucleating agent (ppm)	0	2,000	0	0
MFR (g/10 min)	12.0	12.0	8.1	1.2
Stereoregularity index of ordinary temperature xylene insoluble (%)	96.3	98.9	99.1	99.0
Content of ordinary temperature xylene soluble (wt. %)	25.1	25.0	15.4	34.2
Relaxation time (T1) y (msec)	193	193	133	143
Value of right side of formula (I)	175.2	175.2	132.3	139.9
Flexural modulus (MPa)	760	1,150	1,150	600
-30°C Izod impact strength (kJ/m ²)	3.3	3.2	4.8	91.0
Tensile elongation (%)	≥500	180	≥500	≥500

INDUSTRIAL APPLICABILITY

- The propylene-ethylene block copolymer composition of the present invention is produced at low cost since no additional steps for blending other rubber components are required in the production thereof, and exhibits sufficient stiffness and mechanical strength such as flexural modulus, Izod impact strength and tensile elongation required in automobile exterior parts.

CLAIMS

1. A propylene-ethylene block copolymer composition for automobile exterior parts, comprising a propylene-ethylene block copolymer and a nucleating agent comprising methylenebis(2,4-di-t-butylphenol) acid sodium phosphate which is blended with the propylene-ethylene block copolymer in an amount of 300 to 2,000 ppm upon granulation thereof, and having:
 - 5 (a) a melt flow rate (MFR) of 10 to 18 g/10 min when measured at 230°C under a load of 2.16 kg (21.2N);
 - (b) an ordinary temperature xylene insoluble having a stereoregularity index [mmmm] fraction of 98.9% or higher when measured by ^{13}C -NMR; and
 - 10 (c) an ordinary temperature xylene soluble characterized by:
 - (c-1) having a content of 22 to 28% by weight;
 - (c-2) comprising only a single component with respect to a relaxation time T1 measured by pulse NMR; and
 - 15 (c-3) satisfying the following formula (I):
$$y \leq 0.0014x^3 - 0.0897x^2 - 1.0593x + 231.6 \quad (\text{I})$$
wherein x is an ethylene content (% by weight) measured by ^{13}C -NMR and y is the relaxation time T1 (msec) measured by pulse NMR.
 2. The propylene-ethylene block copolymer composition according to Claim 20 1, wherein said composition has (d) a flexural modulus of 1,000 to 1,500 MPa; (e) a -30°C Izod impact strength of 6 to 8 kJ/m²; and (f) a tensile elongation of 200% or higher.
 3. The propylene-ethylene block copolymer composition according to Claim 25 1, wherein said automobile exterior parts include bumper fascias, splash shields and side moldings .

ABSTRACT OF THE DISCLOSURE

- The present invention provides a propylene-ethylene block copolymer composition for automobile exterior parts having sufficient stiffness and mechanical strength required for the automobile exterior arts, which is
- 5 produced at low cost since no additional steps for blending other rubber components are required in the production thereof. The composition contains methylenebis(2,4-di-t-butylphenol) acid sodium phosphate which is blended with the propylene-ethylene block copolymer in an amount of 300 to 2,000 ppm upon granulation thereof, and has (a) a melt flow rate of 10 to 18 g/10 min when
- 10 measured at 230°C under a load of 2.16 kg (21.2N); (b) an ordinary temperature xylene insoluble having a stereoregularity index [mmmm] fraction of 98.9% or higher when measured by ^{13}C -NMR; and (c) an ordinary temperature xylene soluble characterized by: (c-1) having a content of 22 to 28% by weight; (c-2) comprising only a single component with respect to a relaxation time T1
- 15 measured by pulse NMR; and (c-3) satisfying the following formula (I):

$$y \leq 0.0014x^3 - 0.0897x^2 - 1.0593x + 231.6 \quad (\text{I})$$

wherein x is an ethylene content (% by weight) measured by ^{13}C -NMR and y is the relaxation time T1 (msec) measured by pulse NMR.

Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関する請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者（下記の名称が複数の場合）であると信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled,

上記登録の明細書は、

- 本書に添付されています。
 - ____月____日に提出され、米国出願番号または特許協定条約国際出願番号を_____とし、
(該当する場合) _____に訂正されました。

the specification of which

- is attached hereto.
 was filed on March 29, 2000
as United States Application Number or
PCT International Application Number
PCT/JP00/01919 and was amended on
(if applicable).

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

Japanese Language Declaration

(日本語宣言書)

私は、米国法典第35編119条 (a) - (d) 項又は365条 (b) 項に基づき下記の、米国以外の國の少なくとも一ヵ国を指定している特許協力条約365 (a) 項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

Prior Foreign Application(s)

国外での先行出願

091644/1999	Japan	31/03/99	Priority Claimed 優先権主張
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願年月日)	<input checked="" type="checkbox"/> Yes はい <input type="checkbox"/> No いいえ
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願年月日)	<input type="checkbox"/> Yes はい <input checked="" type="checkbox"/> No いいえ

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I hereby claim foreign priority under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Priority Claimed

<input checked="" type="checkbox"/>	Yes はい	<input type="checkbox"/>	No いいえ
<input type="checkbox"/>	Yes はい	<input checked="" type="checkbox"/>	No いいえ

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

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I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

(Application No.) (出願番号)	(Filing Date) (出願日)
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(Status: Patented, Pending, Abandoned)
(現況 : 特許可済、係属中、放棄済)

(Application No.) (出願番号)	(Filing Date) (出願日)
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(Status: Patented, Pending, Abandoned)
(現況 : 特許可済、係属中、放棄済)

私は、私自信の知識に基づいて本宣言書で私が行なう表明が真実であり、かつ私の入手した情報と私の信じるところに基づく表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第18編第1001条に基づき、罰金または拘禁、もしくはその両方により处罚されること、そしてそのような故意による虚偽の声明を行なえば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに上記のことごとく宣誓を致します。

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Japanese Language Declaration

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委任状：私は下記の発明者として、本出願に関する一切の手続きを米特許商標局に対して遂行する弁理士または代理人として、下記の者を指名いたします。
(弁護士、または代理人の指名及び登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: (list name and registration number)

Norman F. Oblon, Reg. No. 24,618; Marvin J. Spivak, Reg. No. 24,913; C. Irvin McClelland, Reg. No. 21,124; Gregory J. Maier, Reg. No. 25,599; Arthur I. Neustadt, Reg. No. 24,854; Richard D. Kelly, Reg. No. 27,757; James D. Hamilton, Reg. No. 26,421; Eckhard H. Kuesters, Reg. No. 28,870; Robert T. Pous, Reg. No. 29,099; Charles L. Gholtz, Reg. No. 26,395; William E. Beaumont, Reg. No. 30,996; Jean-Paul Lavallee, Reg. No. 31,451; Stephen G. Baxter, Reg. No. 32,884; Richard L. Treanor, Reg. No. 36,379; Steven P. Wehrrouch, Reg. No. 32,829; John T. Goolkasian, Reg. No. 26,142; Richard L. Chinn, Reg. No. 34,305; Steven E. Lipman, Reg. No. 30,011; Carl E. Schlier, Reg. No. 34,426; James J. Kulbaski, Reg. No. 34,648; Richard A. Neifeld, Reg. No. 35,299; J. Derek Mason, Reg. No. 35,270; Surinder Sachar, Reg. No. 34,423; Jeffrey B. McIntyre, Reg. No. 36,867; William T. Enos, Reg. No. 33,128; Michael E. McCabe, Jr., Reg. No. 37,182; Bradley D. Lytle, Reg. No. 40,073; and Michael R. Casey, Reg. No. 40,294, with full powers of substitution and revocation.

書類送付先

Send Correspondence to:

OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P.C.
FOURTH FLOOR
1755 JEFFERSON DAVIS HIGHWAY
ARLINGTON, VIRGINIA 22202 U.S.A.

直接電話連絡先：(名前及び電話番号)

Direct Telephone Calls to: (name and telephone number)
(703) 413-3000

単独発明者または第一の共同発明者の氏名		Full name of sole or first joint inventor <u>Yutaka KOBAYASHI</u>	
発明者の署名	日付	Inventor's signature <u>Yutaka Kobayashi</u> Date Nov. 14, 2000	
住所	Residence Chiba-ken, Japan		
国籍	Citizenship Japan		
郵便の宛先	Post Office Address 1-1, Anesakikaigan, Ichihara-shi,		
	Chiba-ken, Japan <u>JPX</u>		
第二の共同発明者の氏名		Full name of second joint inventor, if any <u>Kenji TANAKA</u>	
第二の共同発明者の署名	日付	Second joint Inventor's signature <u>Kenji Tanaka</u> Date Nov. 14, 2000	
住所	Residence Chiba-ken, Japan <u>JPX</u>		
国籍	Citizenship Japan		
郵便の宛先	Post Office Address 1-1, Anesakikaigan, Ichihara-shi,		
	Chiba-ken, Japan		

(第三以降の共同発明者についても同様に記載し、署名すること)

(Supply similar information and signature for third and subsequent joint inventors.)

Japanese Language Declaration

(日本語宣言書)

300

第三の共同発明者の氏名	Full name of third joint inventor, if any Yoshihisa SAEKI		
第三の共同発明者の署名	日付	Third joint Inventor's signature <i>Yoshihisa SAEKI</i>	Date Nov. 14, 2000
住所	Residence Saitama-ken, Japan		
国籍	Citizenship Japan		
郵便の宛先	Post Office Address c/o Kabushiki Kaisha Honda Gijutsu Kenkyusho 4-1, Chuo 1-chome, Wako-shi, Saitama-ken, Japan		

第四の共同発明者の氏名	Full name of fourth joint inventor, if any		
第四の共同発明者の署名	日付	Fourth joint Inventor's signature	Date
住所	Residence		
国籍	Citizenship		
郵便の宛先	Post Office Address		

第五の共同発明者の氏名	Full name of fifth joint inventor, if any		
第五の共同発明者の署名	日付	Fifth joint Inventor's signature	Date
住所	Residence		
国籍	Citizenship		
郵便の宛先	Post Office Address		

第六の共同発明者の氏名	Full name of sixth joint inventor, if any		
第六の共同発明者の署名	日付	Sixth joint Inventor's signature	Date
住所	Residence		
国籍	Citizenship		
郵便の宛先	Post Office Address		

(第六またはそれ以降の共同発明者に対しても同様な情報および署名を提供すること。)

(Supply similar information and signature for third and subsequent joint inventors.)